

Quasi-one-dimensional disordered systems: fluctuations, transport and interplay

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In a one dimensional lattice thermal fluctuations destroy the long-range order making particles of the lattice move on a scale much larger than the lattice spacing. We discuss the assumption that this motion may be responsible for the transport of localized electrons in a system of weakly coupled chains. The model with diffusing localization sites gives a temperature-independent mobility with a crossover to an activated dependence at high temperature. This prediction is consistent with and might account for experimental results on discotic liquid crystals and certain biopolymers.

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I. INTRODUCTION

In one and two dimensional (D) systems positional correlations diverge with the system size due to thermal fluctuations. Such loss of long-range order, known as Landau-Peierls instability, implies the divergence of the mean square displacement (MSD) of system's structural units. For harmonic lattices this result may be illustrated by direct calculations. Let q_i be a displacement of the i -th atoms in a harmonic chain from its equilibrium position $x_i \sim ia$ where a is the lattice spacing. Assuming that the atoms are distributed as in thermal equilibrium, one can show that the atomic MSD q_i is proportional to the atom's distance from the chain's end,

$$\langle q_i^2 \rangle = \frac{k_B T}{m\omega_0^2} i. \quad (1)$$

Here $\omega_0 = \sqrt{k/m}$, k is the harmonic force constant, and m is the mass of an atom. In a sufficiently long chain the atomic displacement from equilibrium position $\langle q_i^2 \rangle^{1/2}$ may be significantly longer than the equilibrium lattice spacing a . For instance, for the force constant $k \sim 1 \text{ N/m}$, temperature $T \sim 10^2 \text{ K}$, and $i = 10^4$ the equation (1) predict the displacement of order 100 \AA . In the limit of the infinite chain the MSD diverges and atomic motion is unbounded. The loss of long-range order due to Landau-Peierls instability has been observed in X-ray and neutron scattering experiments in many quasi-1D and quasi-2D systems such as liquid crystals and membranes.

The divergence of the MSD does not mean, of course, that the chain is unstable. While the long range order is lost, the short order is preserved. Indeed, the result for the relative displacement of two atoms reads

$$\langle (q_i - q_{i+j})^2 \rangle = \frac{k_B T}{m\omega_0^2} j. \quad (2)$$

According to this equation the distance between two adjacent atoms ($j = 1$) does not depend on the chain's length and deviates from a by a value of order $(k_B T / m\omega_0^2)^{1/2}$, which is normally small.

In recent paper [1] it was suggested that since individual atoms in a 1D lattice are to some extent delocalized, they may serve as temporary vehicles for localized electrons. Consider a system of parallel chains separated by the distance b which is larger than the lattice spacing a . As will be discussed in the following sections, on a long time scale ($\omega_0 t \gg 1$) a tagged atom in a long isolated harmonic chain behaves as a Brownian particle with the diffusion constant

$$D_a = \frac{kT}{2m\omega_0}, \quad (3)$$

which increases linearly with temperature. On the other hand, the diffusion constant D_h for electronic hopping in a static disordered chain depends on temperature exponentially and may be smaller than D_a for sufficiently low T . For instance, for the nearest-neighbor hopping over uncorrelated sites with the Gaussian energy distribution

$$g(\epsilon) = c \exp[-\epsilon^2 / 2\epsilon_0^2], \quad (4)$$

the hopping diffusion constant has the form [2]

$$D_h = \frac{a^2}{2} \nu \exp \left[-a/L - (\epsilon_0 / k_B T)^2 \right] \quad (5)$$

where L is the localization length of the carrier wave function, and ν is the "attempt frequency". For the typical parameter set $k \sim 10 \text{ N/m}$, $\epsilon_0 \sim 0.1 \text{ eV}$, $a \sim 10 \text{ \AA}$, $\nu / \omega_0 \sim 1$, and $\exp(-a/L) \sim 10^{-3}$, one finds that $D_h \ll D_a$ when $\epsilon_0 / k_B T > 1$. This estimation suggests what we call the hitchhiking mechanism of electronic transport: for sufficiently low temperature the hopping mechanism is responsible only for the transport perpendicular to the chains, while along the chains electrons are transported predominantly by mobile localization sites.

To estimate the diffusion constant $D_{||}$ for the lateral transport along the chains due to the hitchhiking mechanism let us assume that the localization is strong and each atom is associated with a mobile localization site whose MSD as a function of time $\langle \Delta q^2(t) \rangle = \langle [q(t) - q(0)]^2 \rangle$ is given. Suppose also that the transition rates for the inter-chain hopping do not strongly fluctuate around a typical value W_{\perp} . Then the lateral motion of an electron

can be considered as a 1D random walk with the time step $\tau \sim 1/W_\perp$ and the length step $l \sim \langle \Delta q^2(\tau) \rangle^{1/2}$. The corresponding diffusion constant is $l^2/2\tau$, which gives

$$D_\parallel = \frac{1}{2} W_\perp \langle \Delta q^2(W_\perp^{-1}) \rangle. \quad (6)$$

If the atomic MSD is diffusive $\langle q^2(t) \rangle \sim 2D_a t$, as in an isolated chain, then D_\parallel does not depend on W_\perp and coincides with the atomic diffusion coefficient

$$D_\parallel = D_a = \frac{k_B T}{2m\omega_0}. \quad (7)$$

The corresponding mobility is temperature independent

$$\mu = \frac{eD_\parallel}{k_B T} = \frac{e}{2m\omega_0} = \frac{e\omega_0}{2k}. \quad (8)$$

For $\omega_0 = 10^{13} \text{ s}^{-1}$ and $k = 10 \text{ N/m}$ this equation gives $\mu \sim 10^{-3} \text{ cm}^2(\text{Vs})^{-1}$, which is consistent with experimental values for columnar liquid crystals and certain conjugated polymers.

Temperature independent mobility was observed in many low-dimensional soft matter systems, in particular in discotic liquid crystals [3] and DNA [4]. It is usually explained in terms of the polaron model [3] or the dynamical disorder models [5]. The model of hitchhiking transport gives an alternative and very simple explanation, which does not involve any adjustable parameters. The model also predicts a crossover to an activated temperature dependence for sufficiently high temperature when the inequality $D_h \ll D_a$ is no longer valid, and the conventional hopping mechanism begins to dominate. Such crossover was reported for charge transport in DNA [4].

The underlying issue of the model is the one of delocalized single-particle dynamics in low-dimensional lattices. The purpose of this paper is to illustrate this issue for two simplest approximations, one of non-interacting chains, and the other of dissipative chains, subjected to the noise and friction forces.

II. LACK OF LONG-RANGE ORDER

Although delocalization of an atom in low-dimensional lattices is a well-known result, it might appear counter-intuitive and is often misinterpreted. For instance, in [6] the divergence of the atomic MSD in a 1D chain is considered as an artifact which arises from the zero-frequency mode in the chain with periodic boundary conditions. Actually, delocalization is the general property of low-dimensional lattices and does not depend on the type of boundary conditions. The question was comprehensively studied by Montroll [7], but the generality of his approach makes it rather complicated. In this section we give a simple derivation of Eq. (1) for a harmonic chain and generalize it for the quantum case.

Consider a harmonic chain of $N + 2$ atoms with fixed ends. Labeling atoms by index $i = 0, 1, \dots, N + 1$ one can

write the Hamiltonian in the form

$$H = \frac{1}{2m} \sum_{i=1}^N p_i^2 + \frac{m\omega_0^2}{2} \sum_{i=1}^{N+1} (q_i - q_{i-1})^2 \quad (9)$$

assuming that displacements for the terminal atoms are zero, $q_0 = q_{N+1} = 0$. The Hamiltonian can be diagonalized by means of a normal mode transformation,

$$q_i = \frac{1}{\sqrt{m}} \sum_{j=1}^N A_{ij} Q_j, \quad p_i = \sqrt{m} \sum_{j=1}^N A_{ij} P_j \quad (10)$$

with normalized eigenvectors

$$A_{ij} = \left(\frac{2}{N+1} \right)^{\frac{1}{2}} \sin \left(\frac{\pi i j}{N+1} \right) \quad (11)$$

which satisfy the orthogonality condition $\sum_{i=1}^N A_{ij} A_{ik} = \delta_{jk}$. In terms of normal coordinates the Hamiltonian assumes the form,

$$H = \frac{1}{2} \sum_{j=1}^N (P_j^2 + \omega_j^2 Q_j^2) \quad (12)$$

where the normal mode frequencies are

$$\omega_j = 2\omega_0 \sin \left[\frac{\pi j}{2(N+1)} \right]. \quad (13)$$

Then the atomic MSD can be written as

$$\langle q_i^2 \rangle = \frac{1}{m} \sum_{jj'} A_{ij} A_{ij'} \langle Q_j Q_{j'} \rangle \quad (14)$$

where the average is taken with respect to the canonical distribution $\rho = Z^{-1} e^{-H/k_B T}$. Since

$$\langle Q_j Q_{j'} \rangle = \delta_{jj'} \frac{k_B T}{\omega_j^2}, \quad \langle P_j P_{j'} \rangle = \delta_{jj'} k_B T, \quad (15)$$

the equation (14) takes the form

$$\langle q_i^2 \rangle = \frac{k_B T}{m} \sum_{j=1}^N \frac{A_{ij}^2}{\omega_j^2}. \quad (16)$$

Inserting the expressions for A_{ij} and ω_j , and introducing the new variable

$$\theta_j = \frac{\pi j}{2(N+1)} \quad (17)$$

one obtains

$$\langle q_i^2 \rangle = \frac{k_B T}{4m\omega_0^2} \frac{2}{N+1} \sum_{j=1}^N \frac{\sin^2(2i\theta_j)}{\sin^2 \theta_j}. \quad (18)$$

Since $\Delta\theta_j = \theta_{j+1} - \theta_j = \pi/2(N+1)$, the sum in the above expression can be converted in the limit $N \rightarrow \infty$ into the integral as follows

$$\langle q_i^2 \rangle = \frac{k_B T}{m\omega_0^2} \frac{1}{\pi} \int_0^{\frac{\pi}{2}} \frac{\sin^2(2i\theta)}{\sin^2 \theta} d\theta \quad (19)$$

which eventually gives

$$\langle q_i^2 \rangle = \frac{k_B T}{m \omega_0^2} i = \frac{k_B T}{k} i. \quad (20)$$

The equation (2) can be derived in a similar way.

In fact, the result (20) can be obtained without the normal mode transformation, but using instead new coordinates $\delta_i = q_i - q_{i-1}$ and noticing that $q_i = \sum_{k=1}^i \delta_k$. This formula is the manifestation of cumulative nature of deviation from equilibrium in 1D systems. Another comment concerns the equations (16), the summation of which gives

$$\sum_i \langle q_i^2 \rangle = \frac{k_B T}{m} \sum_j \omega_j^{-2}. \quad (21)$$

This relation does not involve A_{ij} and is believed to be a general result [8].

The shortest way to generalize the result (20) for the quantum case is to use the quantum mechanical formula for the average energy of the oscillator, corresponding to a j -th mode

$$\langle E_j \rangle = \frac{\hbar \omega_j}{2} \coth \left(\frac{\hbar \omega_j}{2 k_B T} \right). \quad (22)$$

Since the average potential and kinetic energies are equal,

$$\frac{\omega_j^2}{2} \langle Q_j^2 \rangle = \frac{1}{2} \langle P_j^2 \rangle = \frac{1}{2} \langle E_j \rangle = \frac{\hbar \omega_j}{4} \coth \left(\frac{\hbar \omega_j}{2 k_B T} \right)$$

one obtains

$$\langle Q_j Q_{j'} \rangle = \delta_{jj'} \frac{\hbar}{2 \omega_j} \coth \left(\frac{\hbar \omega_j}{2 k_B T} \right). \quad (23)$$

Substitution of this result into (14) gives

$$\langle q_i^2 \rangle = \frac{\hbar}{m \omega_0} \frac{1}{\pi} \int_0^{\frac{\pi}{2}} \frac{\sin^2(2i\theta)}{\sin \theta} \coth \left(\frac{\hbar \omega_0 \sin \theta}{k_B T} \right) d\theta. \quad (24)$$

In the high temperature limit, $\hbar \omega_0 / k_B T \ll 1$, one can use the approximation $\coth(x) \approx 1/x$, which leads to the classical result (20).

For the ultimate quantum case $T = 0$, $\coth(x)$ goes to one, and the MSD takes the form

$$\langle q_i^2 \rangle = \frac{\hbar}{m \omega_0} \frac{1}{\pi} \int_0^{\frac{\pi}{2}} \frac{\sin^2(2i\theta)}{\sin \theta} d\theta \quad (25)$$

where the integral increases with i logarithmically

$$\int_0^{\frac{\pi}{2}} \frac{\sin^2(2i\theta)}{\sin \theta} d\theta \sim \frac{1}{2} \ln \sqrt{i}. \quad (26)$$

Thus the MSD due to quantum zero-point fluctuations reads as follows,

$$\langle q_i^2 \rangle \sim \frac{\hbar}{2 \pi m \omega_0} \ln i. \quad (27)$$

For $\omega_0 \sim 10^{12} \text{ s}^{-1}$, $m \sim 10^{-27} \text{ kg}$ (proton), and $\ln i \sim 1$, the above equation gives $\langle q_i^2 \rangle^{1/2} \sim 1 \text{ \AA}$.

III. DYNAMICS

With the delocalized character of atomic motion in a long chain established, let us consider the question about the dynamics of this motion. It can be conveniently described in terms of a velocity correlation function

$$C(t_1, t_2) = \langle v(t_1) v(t_2) \rangle. \quad (28)$$

For instance, integrating $C(t_1, t_2)$ one obtains the MSD $\langle \Delta q^2(t) \rangle = \langle [q(t) - q(0)]^2 \rangle$ of an atom:

$$\langle \Delta q^2(t) \rangle = \int_0^t dt_2 \int_0^{t_2} dt_1 C(t_1, t_2). \quad (29)$$

Using stationarity of the process $v(t)$, $C(t_1, t_2) \equiv C(t_2 - t_1)$ and integrating by parts, one gets

$$\langle \Delta q^2(t) \rangle = 2 \int_0^t d\tau (t - \tau) C(\tau). \quad (30)$$

Next, one can show [1] that the Laplace-Fourier transform of the velocity correlation function $\tilde{C}(\omega) = \int_0^\infty dt e^{-i\omega t} C(t)$ determines the dynamical mobility $\mu(\omega)$ of a charged atom,

$$\mu(\omega) = \frac{e}{k_B T} \tilde{C}(\omega). \quad (31)$$

For an isolated chain, $C(t)$ is a Bessel function

$$C(t) = \frac{k_B T}{m} J_0(2\omega_0 t). \quad (32)$$

This result can be obtained using the normal mode transformation of the previous section. Indeed, since

$$v_i(t) = \frac{1}{\sqrt{m}} \sum_{j=1}^N A_{ij} P_j(t) \quad (33)$$

and

$$P_j(t) = P_j(0) \cos \omega_j t - \omega_j Q_j(0) \sin \omega_j t, \quad (34)$$

one obtains for the correlation $C_i(t) = \langle v_i(0) v_i(t) \rangle$

$$C_i(t) = \frac{1}{m} \sum_{j,k=1}^N A_{ij} A_{ik} \langle P_j(0) P_k(0) \rangle \cos \omega_j t. \quad (35)$$

Assuming that initial distribution of coordinates and momenta is canonical, one gets $\langle P_j(0) P_k(0) \rangle = \delta_{jk} k_B T$. Then

$$C_i(t) = \frac{k_B T}{m} \sum_{j=1}^N A_{ij}^2 \cos \omega_j t. \quad (36)$$

Substituting the explicit expressions for A_{ij} and ω_j and converting the sum into an integral one obtains

$$C_i(t) = \frac{k_B T}{m} \frac{4}{\pi} \int_0^{\pi/2} d\theta \sin(2i\theta) \cos(2\omega_0 \sin \theta), \quad (37)$$

or

$$C_i(t) = \frac{k_B T}{m} \left\{ J_0(2\omega_0 t) - J_{4i}(2\omega_0 t) \right\}. \quad (38)$$

For large i the term with J_{4i} may be neglected, and one recovers the result (32).

Using (30) and (32) one obtains for the MSD

$$\langle \Delta q^2(t) \rangle = 2D_a t - 2D_a t J_1(2\omega_0 t), \quad (39)$$

where the diffusion coefficient is

$$D_a = \int_0^\infty dt C(t) = \frac{k_B T}{2m\omega_0}. \quad (40)$$

For long time, $\omega_0 t \gg 1$, the first term in (39) dominates, so that the atomic motion is diffusive, $\langle \Delta q^2(t) \rangle \sim 2D_a t$. This result was first discussed by Rubin [9]. In the same limit the mobility is purely real and does not depend on frequency, $\mu(\omega) \approx e/2\omega_0 m$.

IV. DISSIPATIVE CHAINS

In previous sections the interaction of chains has been neglected which is very likely to be an over-simplification. The problem of dynamics of interacting chains may be considered as a generalization of the Frenkel-Kontorova model about a chain in an external spatially periodic potential field [10]. In quasi-1D systems this periodic potential is created by adjacent chains and is not static, which makes the problem very difficult [11]. It was suggested in [1] that some insight can be achieved by modelling the chains interaction using the Langevin approach. Namely, one may assume that the force exerted on an atom by adjacent chains can be written as the sum of a regular dissipative force linear in the atom's velocity, $-\gamma \dot{q}_i$, and a fluctuating term $\xi_i(t)$. With this assumption the equation of motion of the atoms takes the form

$$m\ddot{q}_i(t) = k(q_{i-1} + q_{i+1} - 2q_i) - \gamma \dot{q}_i(t) + \xi_i(t). \quad (41)$$

Let us treat the fluctuating term $\xi_i(t)$ as a zero centered white noise which is not correlated for different atoms and related to the friction constant γ through the conventional fluctuation-dissipation relation,

$$\langle \xi(t) \rangle = 0, \quad \langle \xi_i(0) \xi_k(t) \rangle = 2k_B T \gamma \delta_{ik} \delta(t). \quad (42)$$

Such approach is common in polymer physics to describe the polymer-solvent interaction. In that case the friction constant γ can be expressed in terms of the solvent viscosity. In our model, the dissipation and fluctuating terms describes interaction with other chains, and γ is an adjustable parameter.

Let us find the velocity correlation function of an atom for the model of dissipative chains. Referring again to the normal mode transformation (10), the inverse has the form

$$Q_j = \sqrt{m} \sum_{i=1}^N A_{ij} q_i, \quad P_j = \frac{1}{\sqrt{m}} \sum_{i=1}^N A_{ij} p_i. \quad (43)$$

Then from the above equation of motion one finds the equation for $Q_j(t)$

$$\ddot{Q}_j(t) = -\omega_j^2 Q_j(t) - \lambda \dot{Q}_j(t) + \eta_j(t). \quad (44)$$

Here

$$\eta_j(t) = \frac{1}{\sqrt{m}} \sum_{i=1}^N A_{ij} \xi_i(t) \quad (45)$$

has a meaning of the random force for a normal mode Q_j , and $\lambda = \gamma/m$ is the inverse velocity relaxation time for an atom. Using the method of Laplace transform the solution of (44) can be written as

$$Q_j(t) = a_j(t) Q_j(0) + b_j(t) \dot{Q}_j(0) + \int_0^t d\tau b_j(t-\tau) \eta_j(\tau). \quad (46)$$

The explicit form of the functions $a_j(t)$ and $b_j(t)$ depends on the sign of the difference $\omega_j - \lambda/2$. We consider here only the case of an over-damped chain when $\lambda > 2\omega_0$ (and therefore $\lambda > 2\omega_j$ for any normal mode j). In this case

$$a_j(t) = e^{-\frac{\lambda}{2}t} \left\{ \frac{\lambda}{2\Omega_j} \sinh \Omega_j t + \cosh \Omega_j t \right\}, \quad (47)$$

$$b_j(t) = \frac{1}{\Omega_j} e^{-\frac{\lambda}{2}t} \sinh \Omega_j t, \quad (48)$$

where $\Omega_j = \sqrt{\lambda^2/4 - \omega_j^2}$. Then for the velocity correlation function $C(t) = \langle \dot{q}_i(t) \dot{q}_i(0) \rangle$ one gets

$$C(t) = \frac{k_B T}{m} \sum_j A_{ij}^2 \dot{b}_j(t). \quad (49)$$

For the very strong dumping $\lambda \gg \omega_0$, $\dot{b}_j(t)$ can be approximated as

$$\dot{b}_j(t) = \exp(-\lambda t) - (\omega_j/\lambda)^2 \exp(-\omega_j^2 t/\lambda). \quad (50)$$

Transforming the sum (49) into an integral one obtains the velocity correlation function in the form

$$C(t) = \frac{k_B T}{m} e^{-\lambda t} + \frac{k_B T}{m\lambda} \frac{d}{dt} \left\{ e^{-\alpha t} I_0(\alpha t) \right\} \quad (51)$$

where $I_0(x)$ is the modified Bessel function, and $\alpha = 2\omega_0^2/\lambda$. Note that $\alpha \ll \lambda$.

Substituting the above expression for $C(t)$ into (30) one obtains

$$\langle \Delta q^2(t) \rangle = \frac{2k_B T}{\gamma} t e^{\alpha t} \left\{ I_0(\alpha t) + I_1(\alpha t) \right\}. \quad (52)$$

For long time, $t \gg \alpha^{-1}$, this expression gives sub-diffusive behavior

$$\langle \Delta q^2(t) \rangle \sim 2F\sqrt{t} \quad (53)$$

with the mobility factor

$$F = \frac{k_B T}{\omega_0 \sqrt{\pi \gamma m}}. \quad (54)$$

Sub-diffusive motion with the MSD growing as \sqrt{t} is characteristic feature for any form of the single file diffusion when overdamped Brownian particles constrained to move in one dimension and are not allowed to pass each other [12]. Its consequence for the hitchhiking model is that the diffusion constant depends on the inter-chain transition rate W_\perp , as follows from equation (6), $D_\parallel = F\sqrt{W_\perp}$. Note however, that this result holds only for the transport on the time scale much longer than $1/W_\perp$; it does not apply for short chains when inter-chain transitions are negligible.

Another consequence of sub-diffusive transport is that, in contrast to the approximation of non-interacting chains, the dynamical mobility essentially depends on frequency. Using (31) one can find that for $\omega \ll \alpha$ both real and imaginary parts increases with ω as $\omega^{1/2}$, namely

$$\mu(\omega) = \frac{e}{2\omega_0} \sqrt{\frac{\omega}{2m\gamma}} (1 - i). \quad (55)$$

Power frequency dependence $\mu(\omega) \sim \omega^s$ with $0 < s < 1$ is typical for many disordered systems. Note however, that for frequency lower than the inter-chain transition rate, one has to take into account inter-chain hopping of carriers. On this time scale the carrier diffusion is normal $\langle \Delta q^2(t) \rangle \sim t$, and $\mu(\omega)$ is almost frequently independent.

V. CONCLUDING REMARKS

This paper promotes the idea that in quasi-1D systems long-range structural fluctuations may carry localized electrons over a considerable distance and that for sufficiently low temperature this may be the dominating mechanism of charge transport. Most of the results are obtained under very idealized assumptions and can hardly be regarded as anything but toy-model calculations. On the other hand, qualitative predictions of the model seem quite general. For instance, while the character of time dependence for the atomic MSD may depend on many factors, the linear dependence of the MSD on temperature is a general property, which is responsible for a temperature independent hitchhiking mobility.

The assumption of one-dimensional dynamics of the chains is also not essential. In this paper we assumed that the chains are parallel and form strongly anisotropic 2D or 3D crystal. One may argue that the model of hitchhiking electronic transport may be relevant to polymer systems, where monomers move sub-diffusively in three dimensions [13]. Another generalization is the case when the electronic localization length is much larger than the lattice spacing, $L \gg a$. This problem is related to the dynamics of a large cluster [14] rather than of a single particle in a chain.

VI. ACKNOWLEDGEMENT

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